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Chapter 2.0

Concentrations of Sulfur and Nitrogen Species

Air pollutant concentrations sampled in 1998 using the CASTNet filter packs are presented and analyzed in this chapter. These concentrations include SO_2, SO_4^{2-}, HNO_3, NH_4^+, NO_3^-, and total NO_3^-(HNO_3 plus NO_3^- aerosol). In addition, annual and quarterly averages measured at a subset of 34 monitoring sites with the most complete historical record from 1989 through 1998 are presented and analyzed for trends using linear regressions and box plots. The selection of the 34 sites is discussed later in the subsection on 10-year trends.

Quarterly average concentrations were constructed from measured weekly filter concentrations. Quarterly averages were calculated for all sites with at least eight valid weekly values. Annual averages were calculated for all sites with at least three valid quarterly averages.

Concentrations

Sulfur Dioxide

Figure 2-1 presents 1998 annual SO_2 concentrations. Quarterly averages are provided in Appendix A. Higher concentrations in the East were observed during the first quarter. The highest concentrations were measured in proximity to the Ohio River from southern Illinois into Pennsylvania with sharp concentration gradients along the peripheries of the network. The highest SO_2 level in the West was measured at Chiricahua, AZ (CHA467).

Particulate Sulfate

Annual average SO_4^{2-} concentrations for 1998 are presented in Figure 2-2. Quarterly averages are given in Appendix A. Higher values were measured in the East during the third quarter with a band of concentrations above 10.0 µg/m^3 from eastern Kentucky into western Virginia. Concentrations were generally higher in 1998 than in 1997 (see Figure 2-10). The annual SO_4^{2-} levels measured throughout the West were generally below 1.5 µg/m^3. During the third quarter, values at several sites exceeded 1.0 µg/m^3.

Nitric Acid

Annual HNO_3 concentrations are shown in Figure 2-3. Quarterly concentrations are given in Appendix A. The eastern data show local geographic variability and sharp gradients along the peripheries of

the network. Values are higher in the third quarter. Concentrations throughout the west are generally lower, except for southern California where HNO_3 equals eastern values.

Particulate Ammonium

Figure 2-4 presents annual averages of NH_4^+ for 1998. Appendix A provides quarterly averages. Eastern concentrations are fairly uniform in the center of the network with gradients along the network peripheries. In general, higher concentrations were measured in agricultural areas, rather than forested areas. Measured NH_4^+ is generally low throughout the West.

Particulate Nitrate

Annual concentrations of NO_3^- aerosols are shown in Figure 2-5. Quarterly average concentrations are given in Appendix A. NO_3^- concentrations are more variable geographically than SO_4^{2-}, with the highest levels measured in agricultural areas in the Midwest (e.g., Illinois and Indiana). Western concentrations are less than 1.0 µg/m^3, except for California.

Total Nitrate

Average annual concentrations of total NO_3^- are depicted in Figure 2-6. Appendix A provides quarterly averages. The concentrations show considerable local geographic variability in the East. The higher values were observed in the Midwest. The pattern of concentrations in the West is similar to HNO_3 with relatively high values over California.

Ten-Year Trends

One of the goals of CASTNet is to assess trends in measured concentrations. Trends analyses have been performed using linear regressions of pollutant concentrations averaged over 34 eastern sites versus year from 1989 through 1998. SO_2 and NO_x emissions for eastern states are also shown on the trends diagrams–SO_2 for trends in sulfur species and NO_x for trends in nitrogen species. Box plots of annual averages over the 10-year period have also been prepared. The 34 sites (Figure 2-7) were selected using criteria similar to those used by EPA (1998) in their annual trends report. Sites with complete data for 8 of the 10 years were selected. Missing quarterly data were interpolated from adjacent quarterly data, e.g., first quarter 1996 data were interpolated from 1995 and 1997 first quarter data. Missing quarterly averages for 1989 or 1998 were assumed equal to adjacent quarterly values. A valid quarterly average was based on eight valid weeks. Annual averages were based on data from four quarters. No trends analyses were performed for western sites based on previous calculations (e.g., CASTNet Deposition Summary Report).

Figure 2-8 shows linear regressions based on annual and quarterly average SO_2 concentrations for the 34-station subset. Annual SO_2 emissions for states east of and including the north-south line of states from Minnesota to Louisiana are also depicted. The figure shows significant reductions in annual and quarterly SO_2 averages. The linear model indicates about a 46% reduction in annual SO_2 levels. The slope of the regression line is smaller than those shown in previous analyses (e.g., 0.68 in the 1997 Annual Report and 0.73 in the Deposition Summary Report). The reduced slope is the result of higher concentrations (and emissions) in 1997 and 1998. The figure also shows a reasonable relationship between the sets of annual emission data and the composite SO_2 concentrations. Quarterly averages have declined significantly over the 10 years. Declines are more pronounced in the two colder quarters when SO_2 emissions are higher and there is little photochemical conversion of SO_2 to SO_4^{2-}.

Figure 2-9 presents trends in the 34-station composite, annual SO_2 levels through a box diagram. The inter-site variability among the 34 sites is shown graphically by the 90th percentile, median, and 10th percentile values for the annual averages. The box plot shows results similar to the regression analysis. Years with lower average SO_2 concentrations also show lower inter-site variability.

Trends in annual and quarterly SO_4^{2-} concentrations are shown in Figure 2-10. Annual SO_2 emissions are also plotted. According to the linear model, annual SO_4^{2-} has declined by about 16% over the 10 years. Third quarter levels show the largest decrease. Concentrations peak in the third quarter because of the enhanced photochemical production of SO_4^{2-} from SO_2. The 1998 third quarter average is higher than those recorded in the previous few years. A box plot of annual SO_4^{2-} levels is given in Figure 2-11.

Linear regressions were also performed on annual and quarterly average HNO_3 concentrations collected from 1989 through 1998. The HNO_3 results for the 34-site composite averages are shown in Figure 2-12. Annual NO_x emissions are also shown. The results show a slight reduction over the 10-year period. A box plot for HNO_3 is given in Figure 2-13.

Linear regressions for annual and quarterly average NH_4^+ concentrations for the 34-site composite database are shown in Figure 2-14. The linear model shows a slight reduction in the 34-station average values over the 10-year period. A box plot for annual average NH_4^+ concentrations is shown in Figure 2-15.

Trends in annual and quarterly particulate NO_3^- concentrations are shown in Figure 2-16. Annual NOx emissions are also plotted. The linear regressions show a slight reduction in concentrations over the 10-year period. A box plot is shown in Figure 2-17.

Trends in annual and quarterly total nitrate concentrations from the 34-station database are shown in Figure 2-18. NO_x emissions are also plotted. No trends are evident from the regression plots. A box plot of annual total nitrate concentrations is given in Figure 2-19.

Figure 2-1. Annual Mean SO_2 Concentrations (µg/m³) for 1998

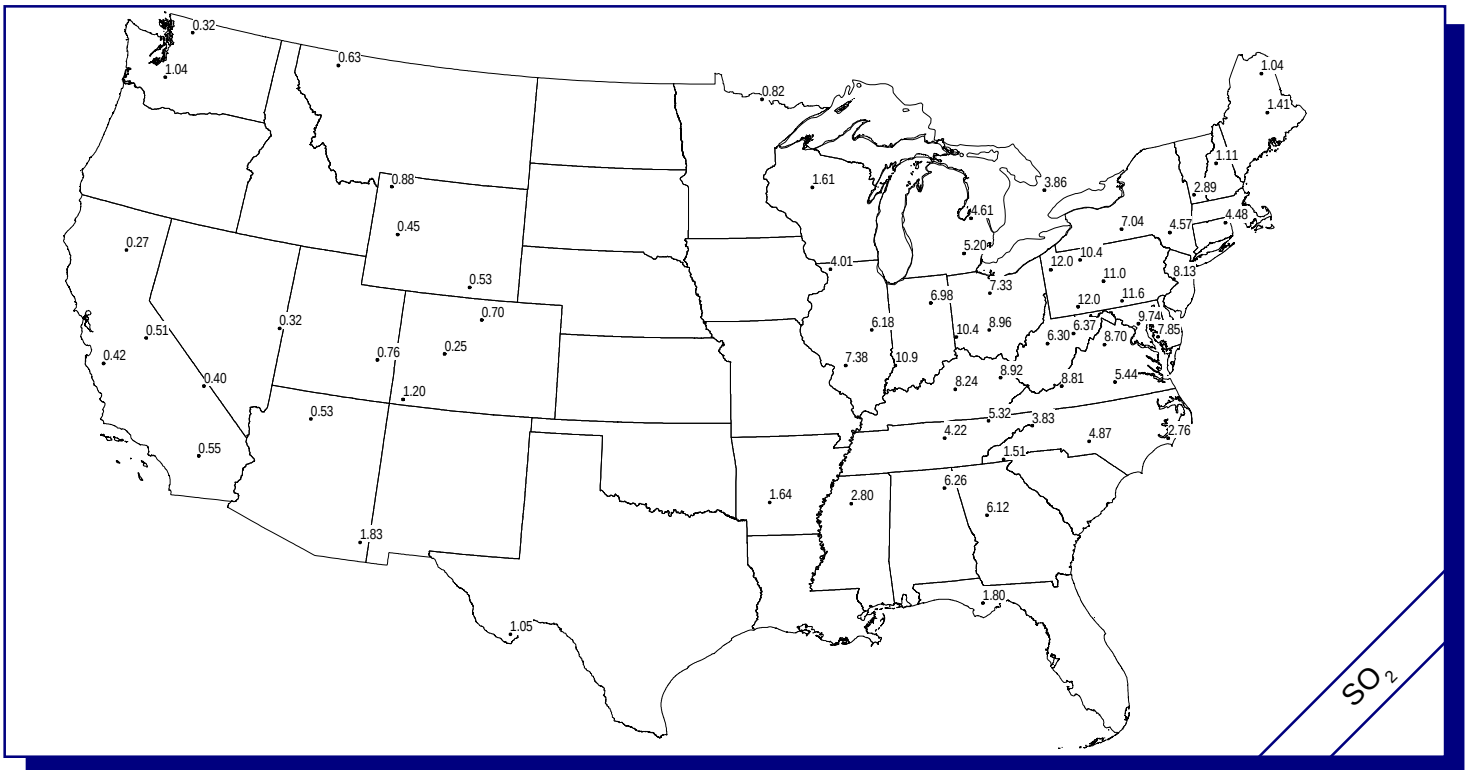


Figure 2-2. Annual Mean Particulate SO_4^{2-} Concentrations (µg/m³) for 1998

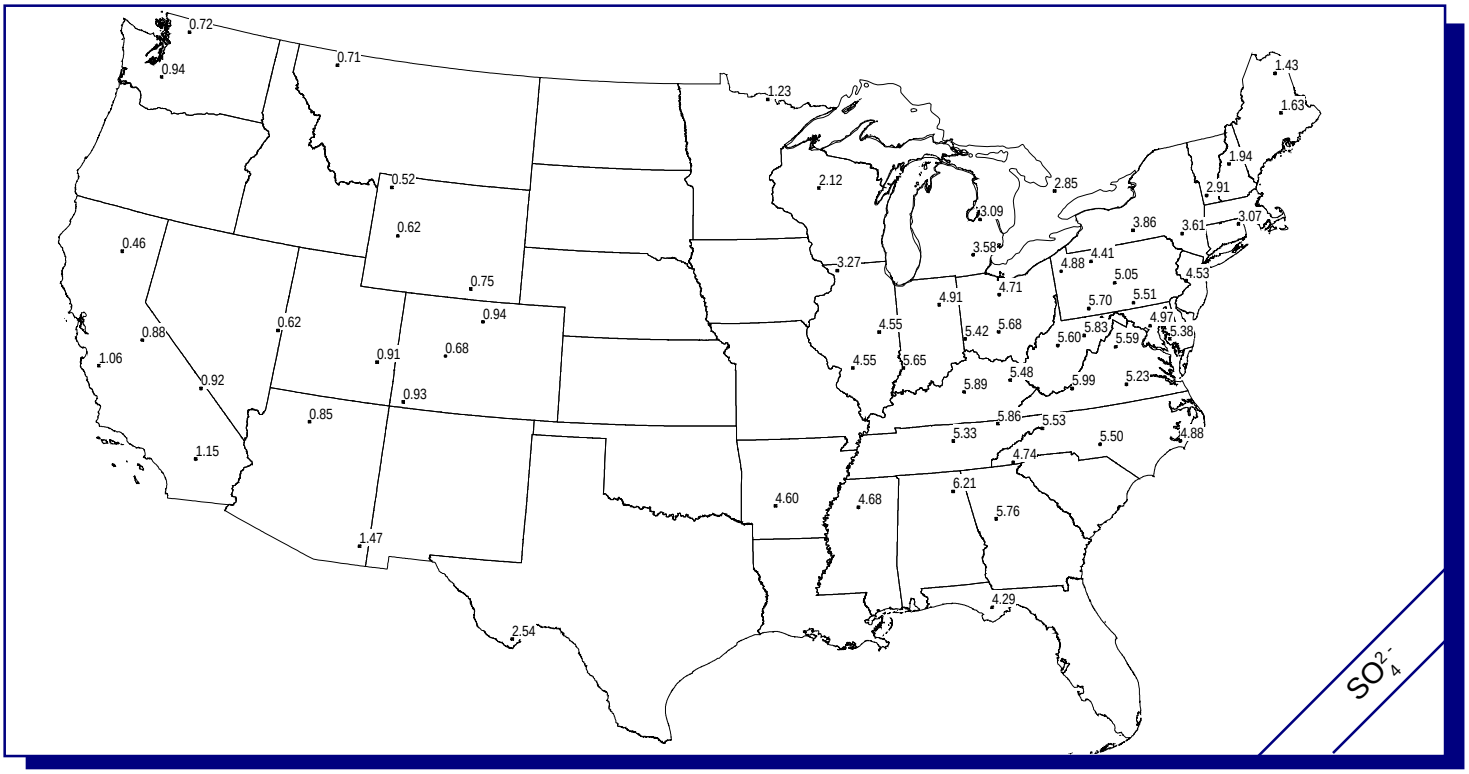


Figure 2-3. Annual Mean HNO_3 Concentrations (µg/m³) for 1998

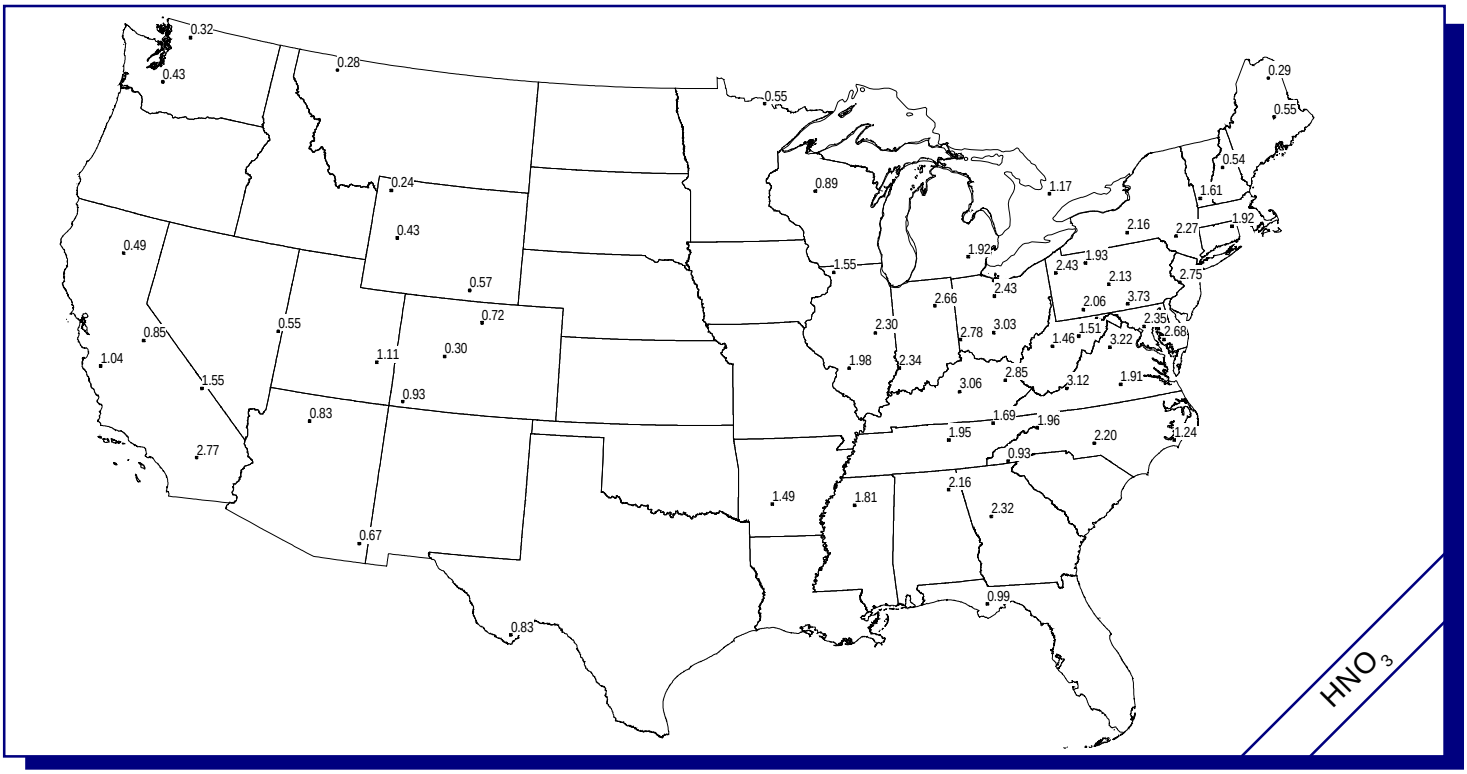


Figure 2-4. Annual Mean Particulate NH_4^+ Concentrations(µg/m³) for 1998

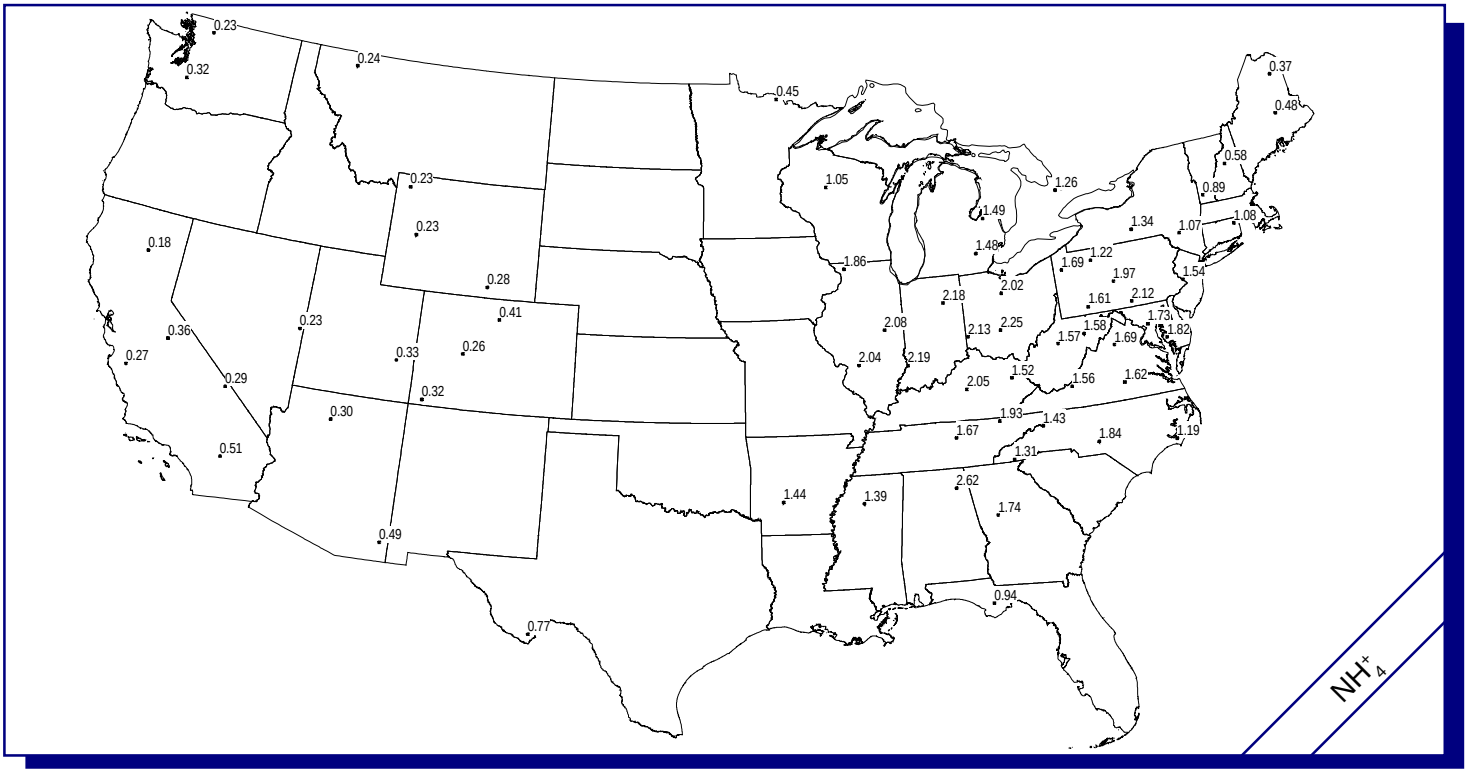


Figure 2-5. Annual Mean Particulate NO_3^- Concentrations (µg/m³) for 1998

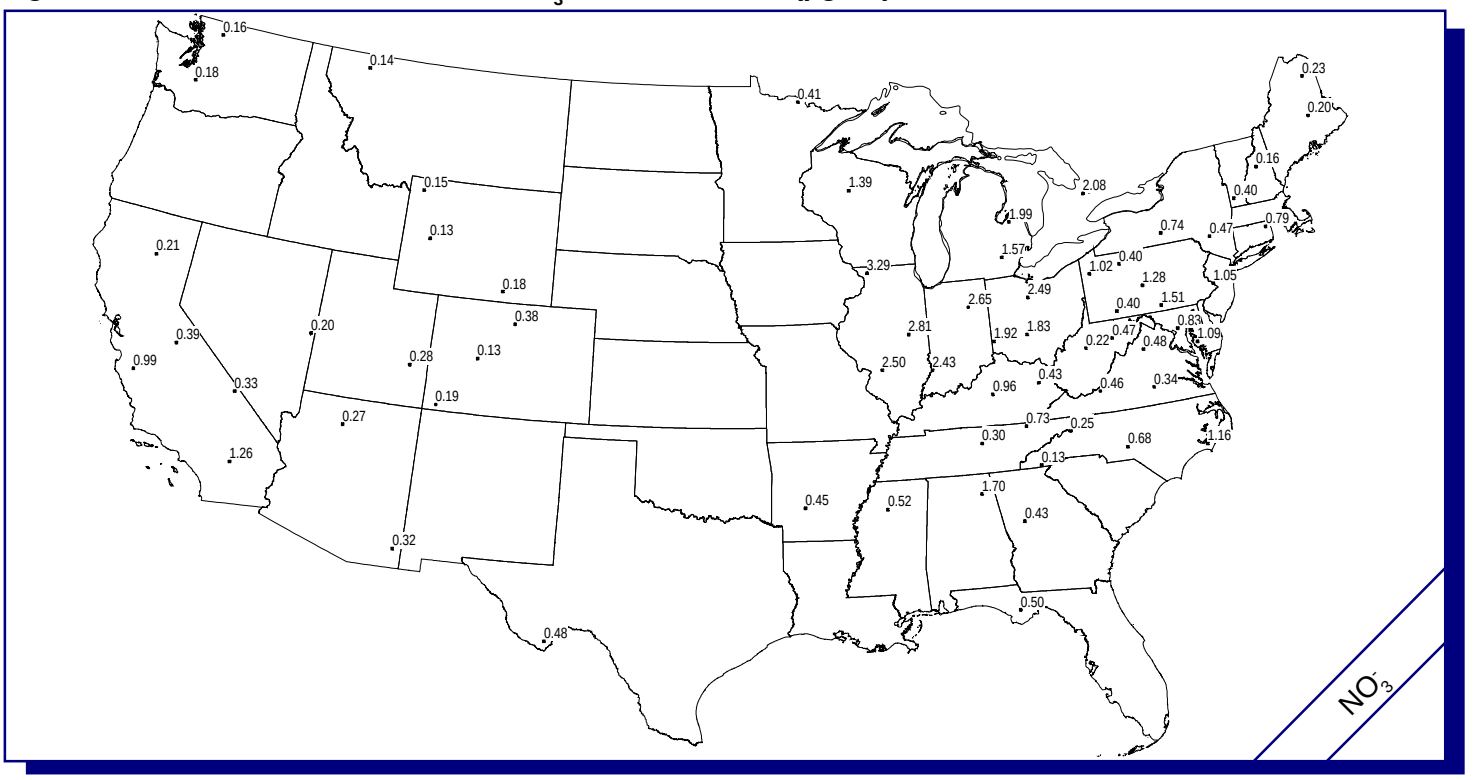


Figure 2-6. Annual Mean Total NO_3^- Concentrations (µg/m³) for 1998

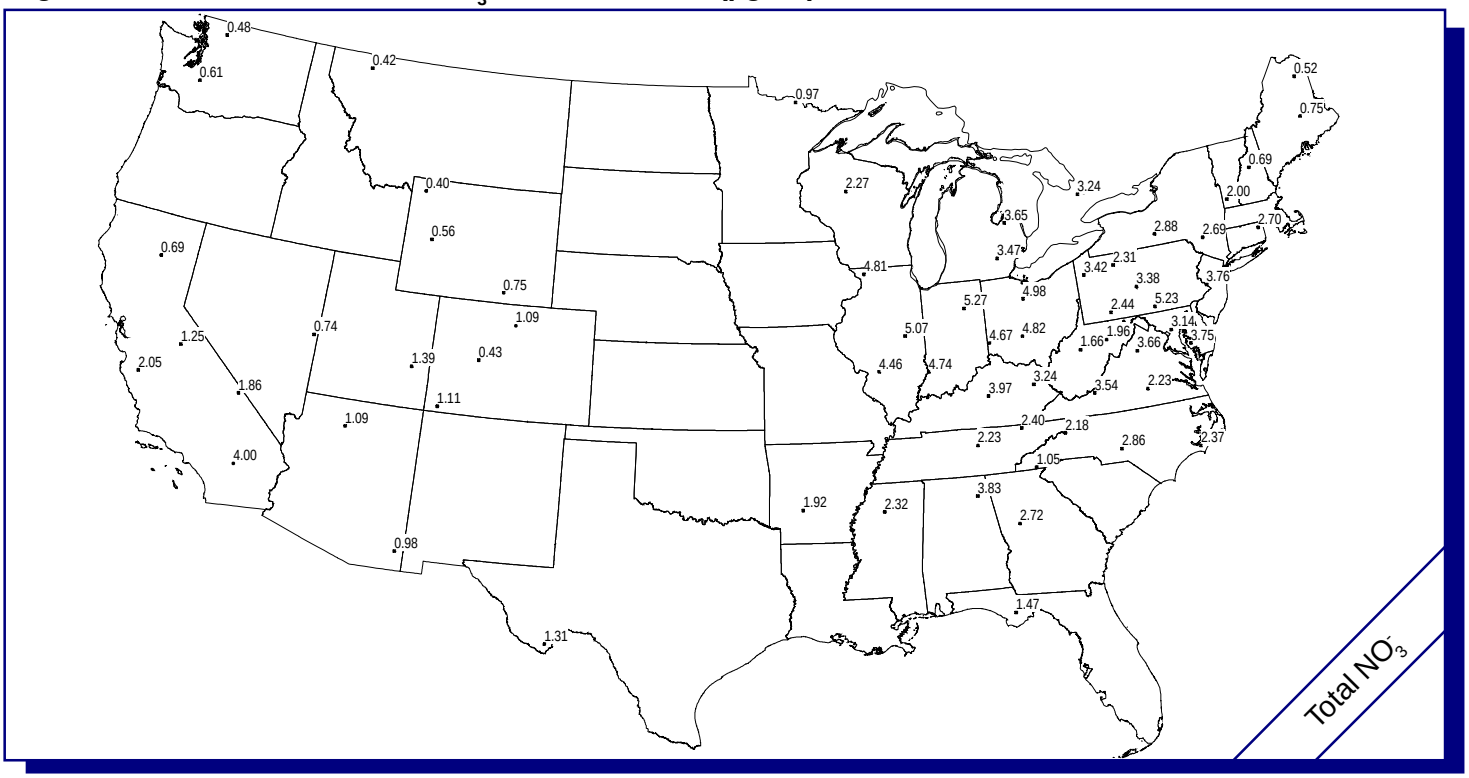


Figure 2-7. Map of 34 Sites Selected for Trends Analysis

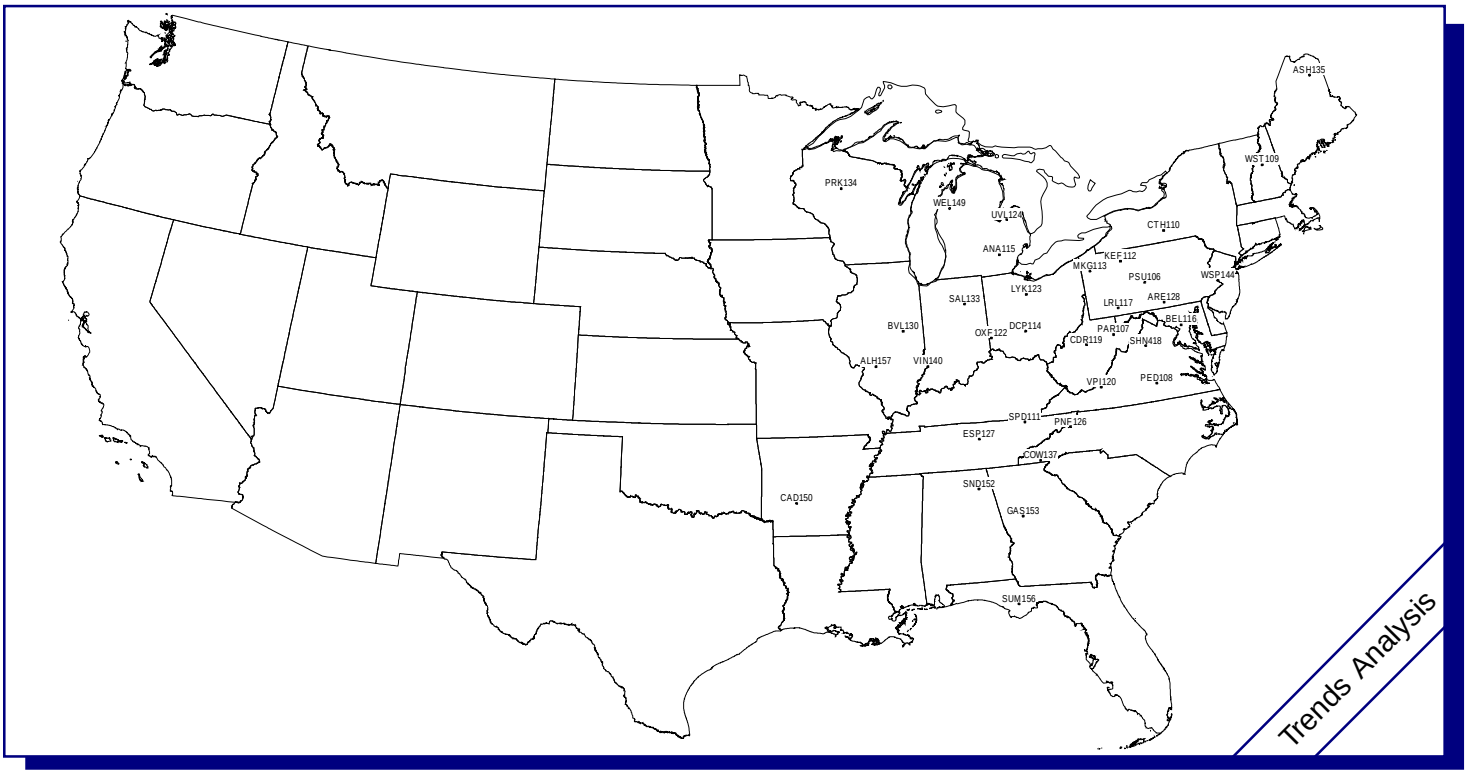
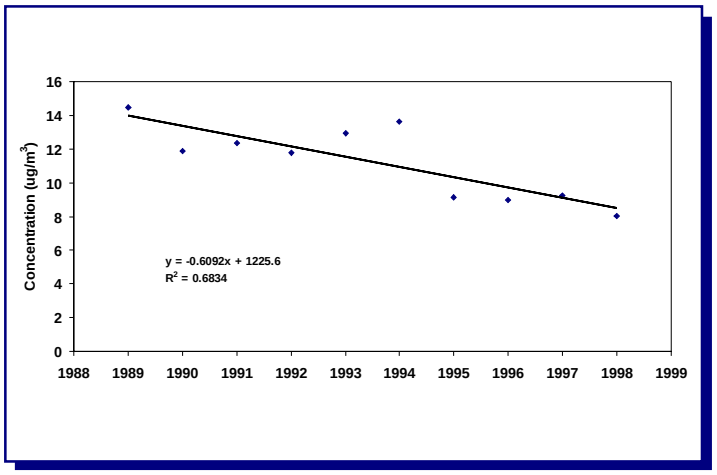
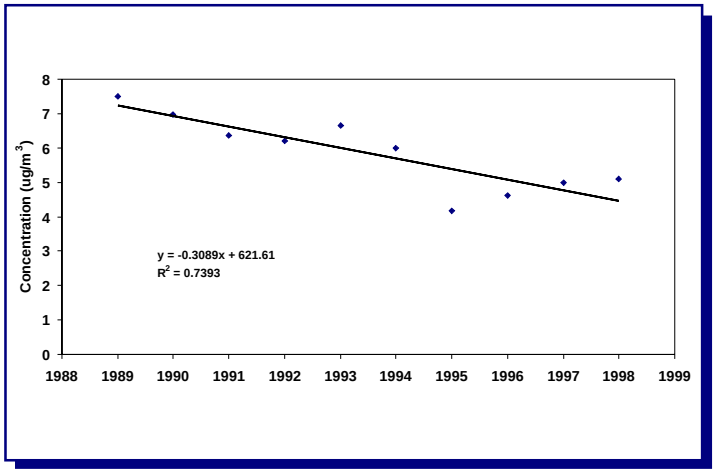


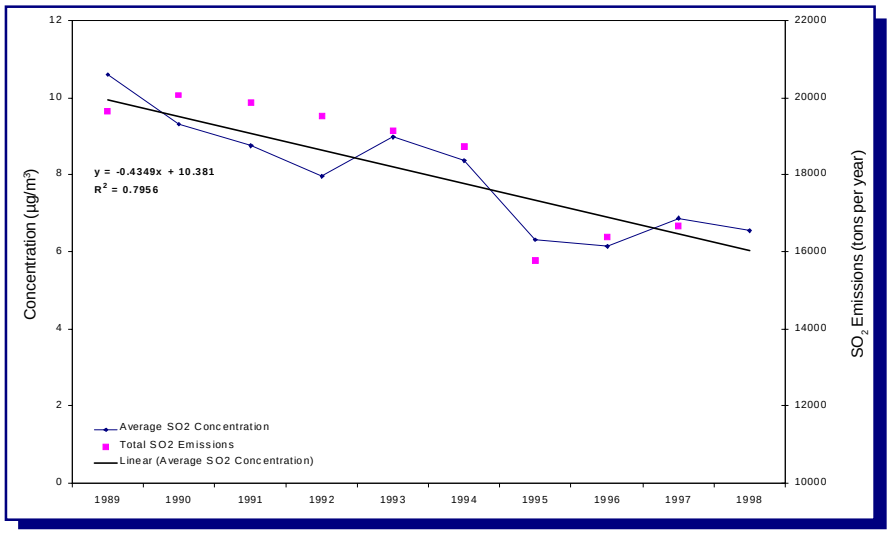
Figure 2-8. Linear Regression Analyses for Annual and Quarterly Average SO_2 Concentrations– Eastern United States



First Quarter

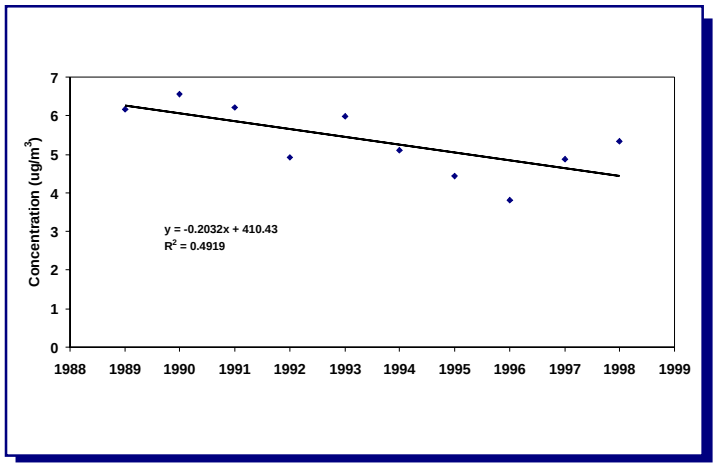


Second Quarter



Annual Average

Third Quarter



Fourth Quarter

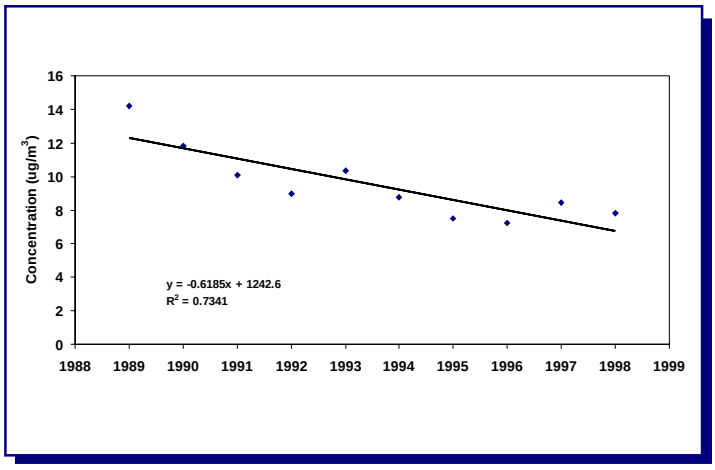


Figure 2-9. Trends in Composite Annual SO_2 Concentrations

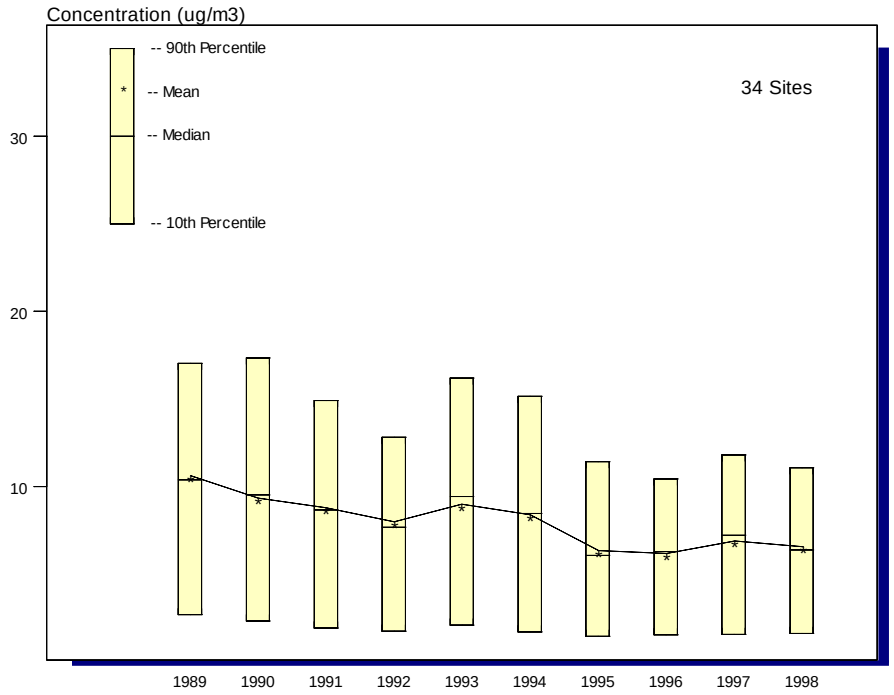
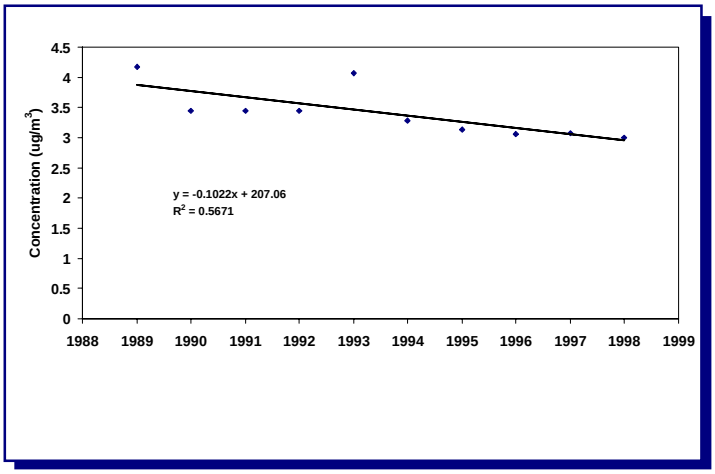
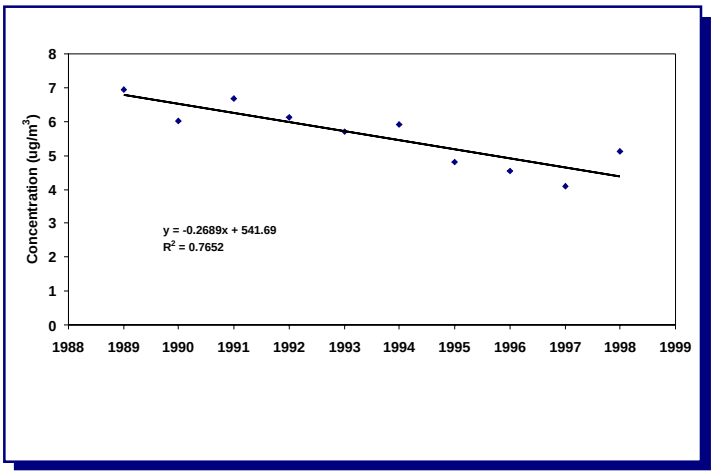


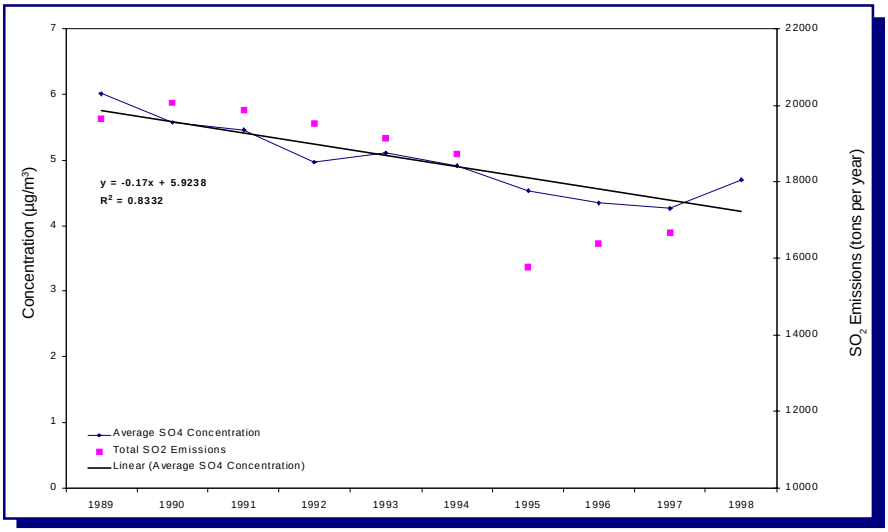
Figure 2-10. Linear Regression Analyses for Annual and Quarterly Average SO_4^{2-} Concentrations– Eastern United States



First Quarter

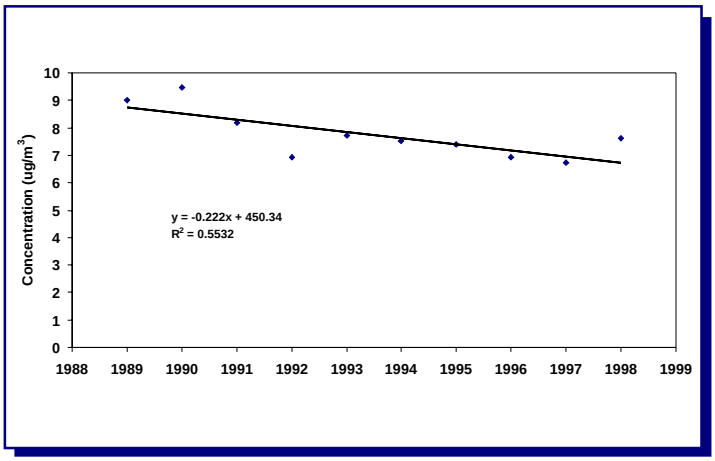


Second Quarter



Annual Average

Third Quarter



Fourth Quarter

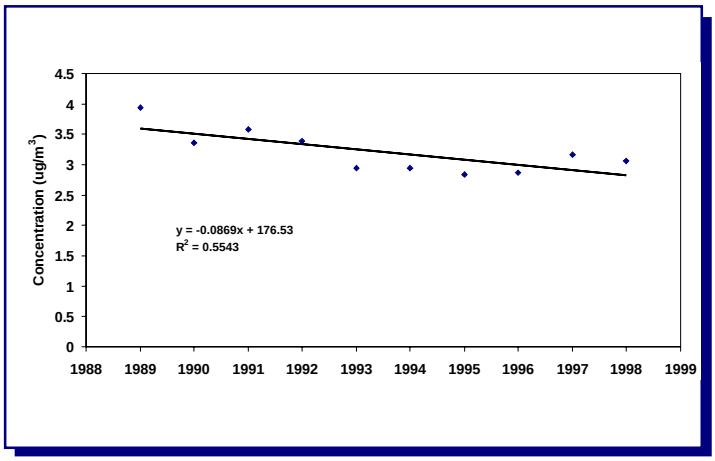


Figure 2-11. Trends in Composite Annual SO_4^{2-} Concentrations

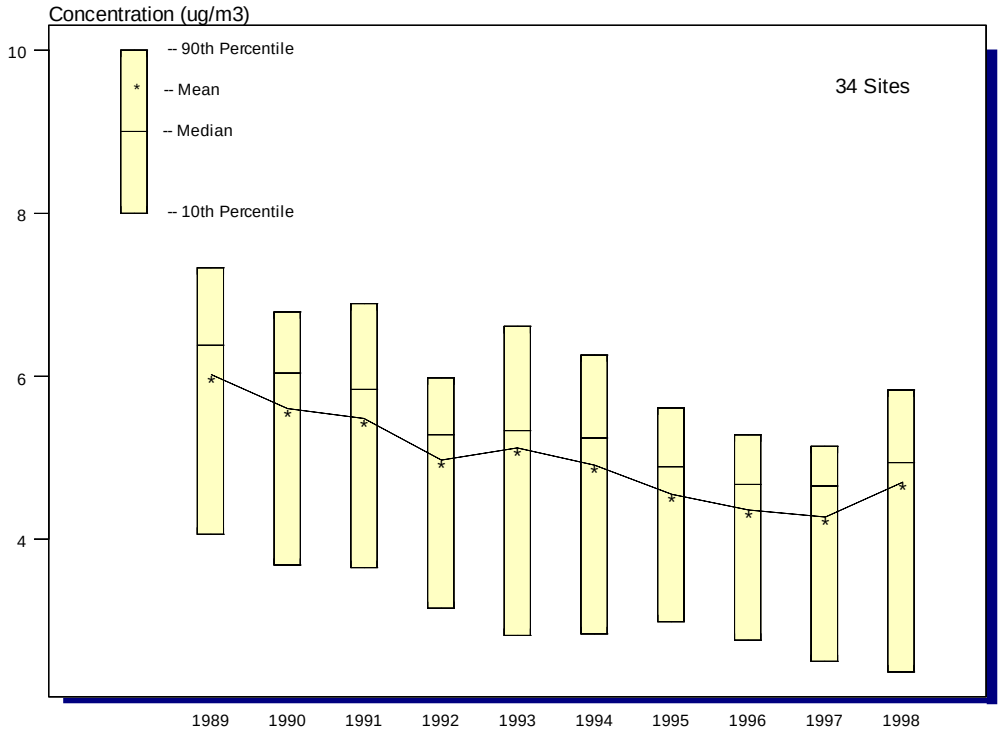
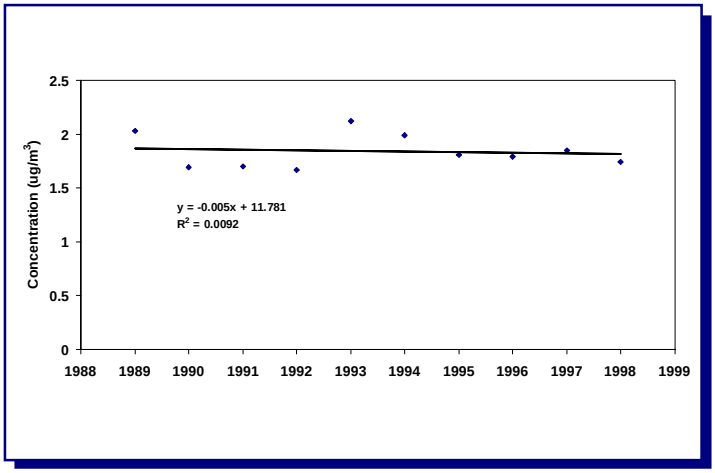
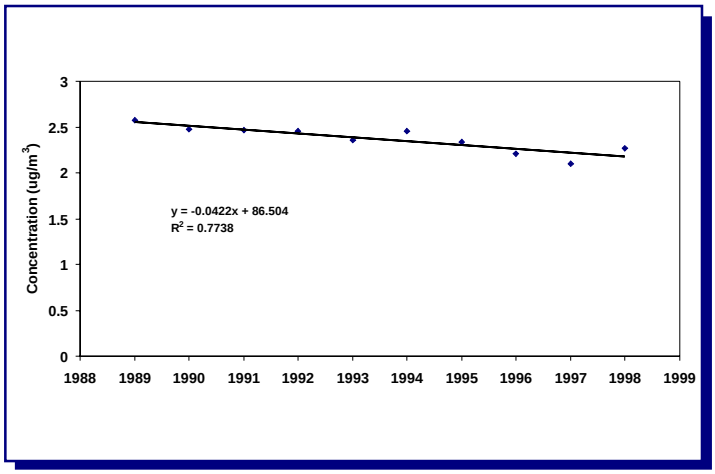


Figure 2-12. Linear Regression Analyses for Annual and Quarterly Average HNO_3 Concentrations– Eastern United States



First Quarter



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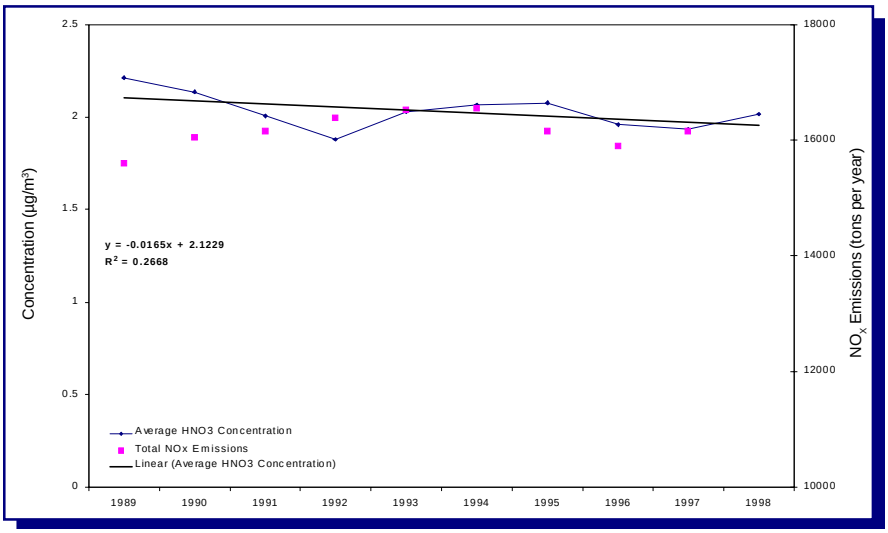
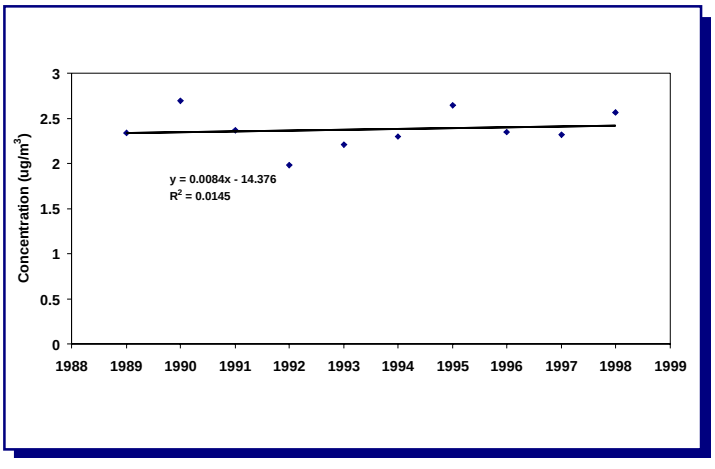


Annual Average

Third Quarter



Fourth Quarter

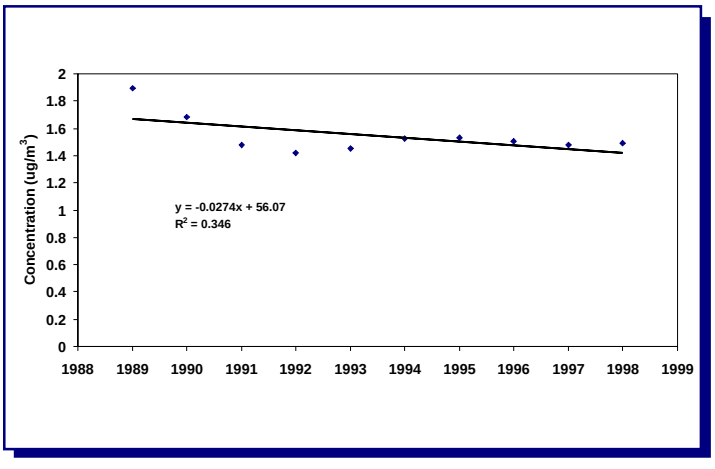


Figure 2-13. Trends in Composite Annual HNO_3 Concentrations

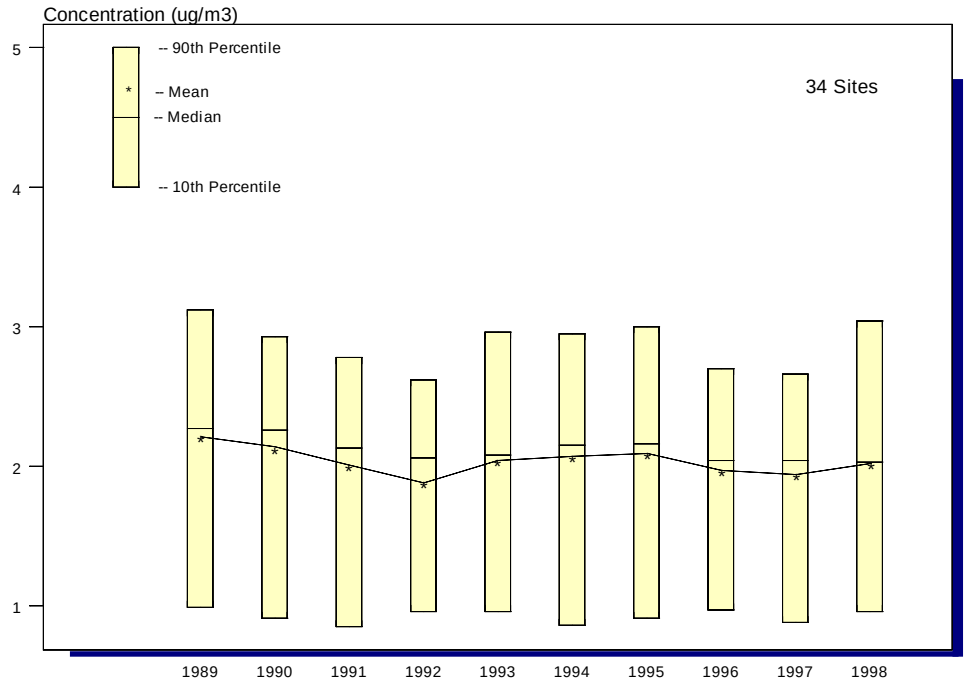
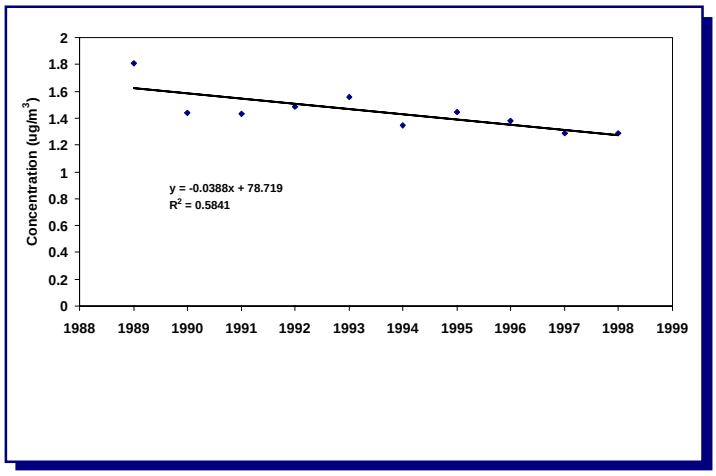
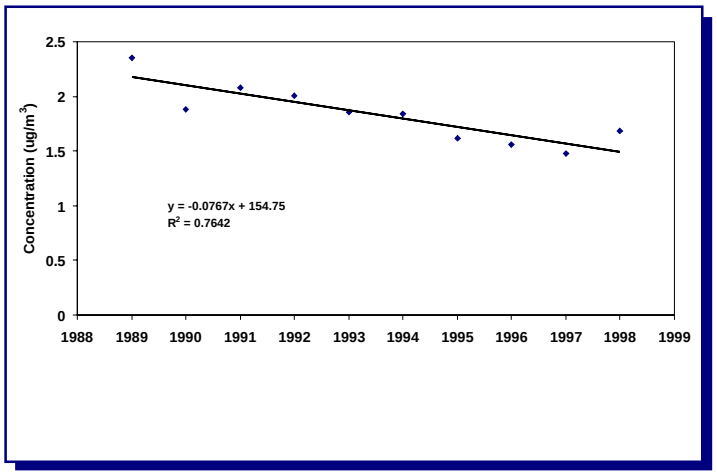


Figure 2-14. Linear Regression Analyses for Annual and Quarterly Average NH_4^+ Concentrations– Eastern United States



First Quarter



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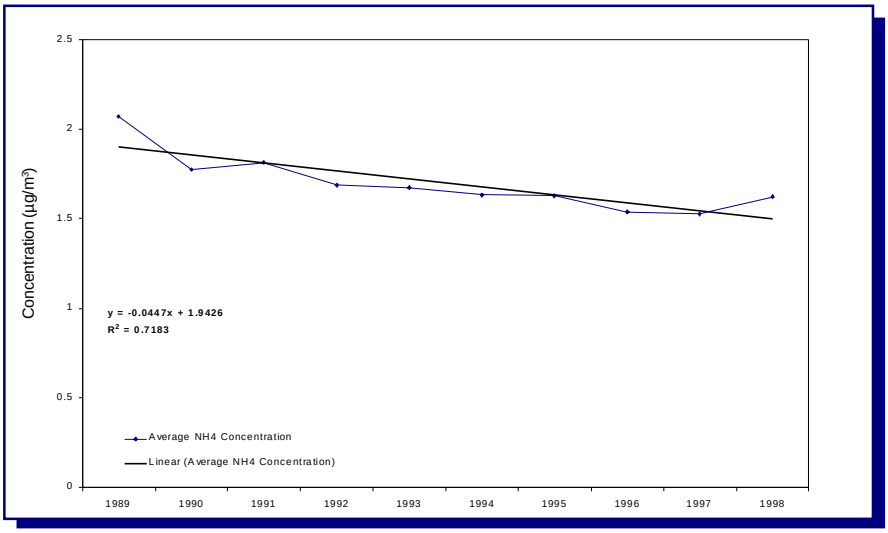
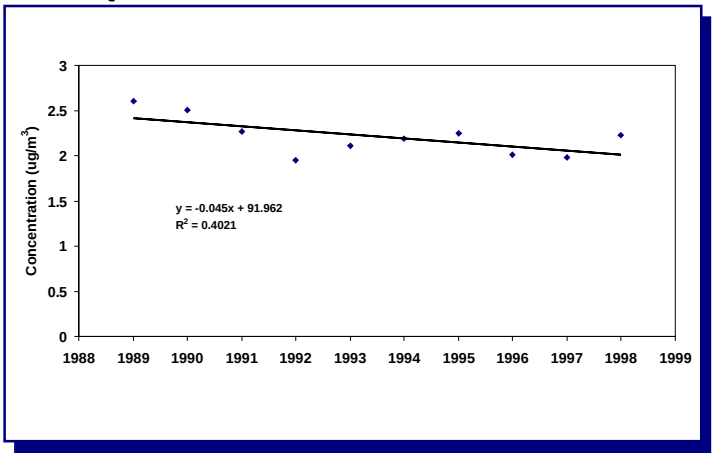


Annual Average

Third Quarter



Fourth Quarter

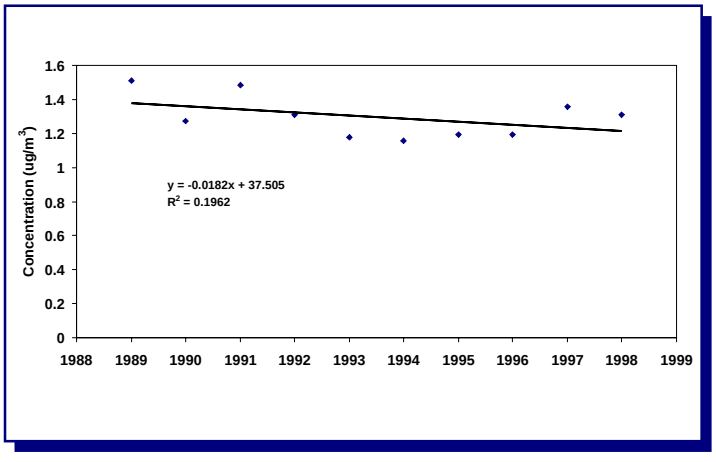


Figure 2-15. Trends in Composite Annual NH_4^+ Concentrations

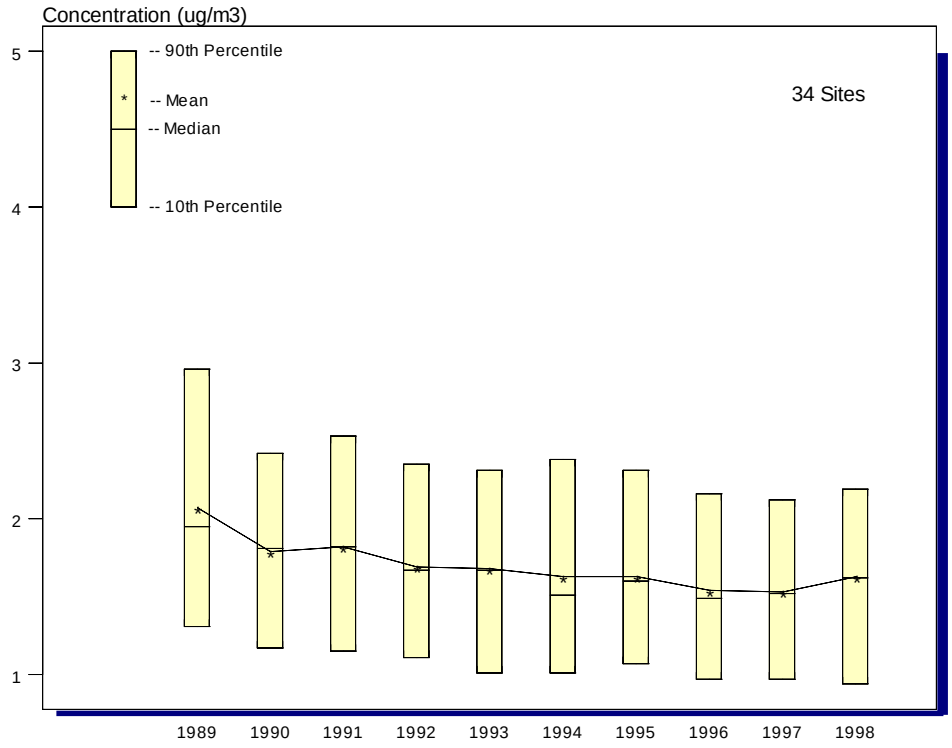
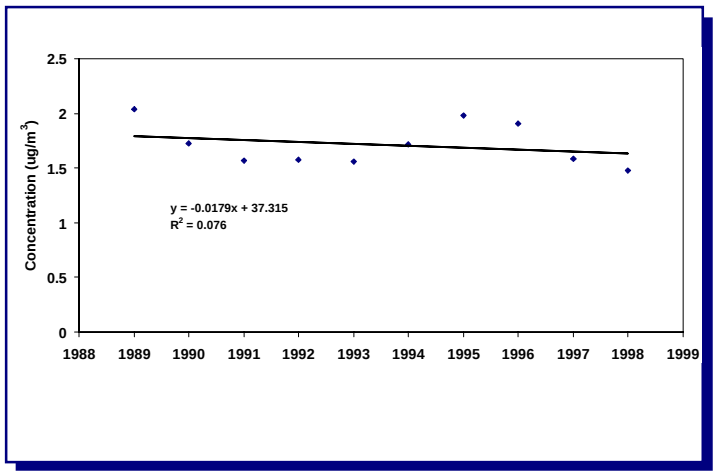
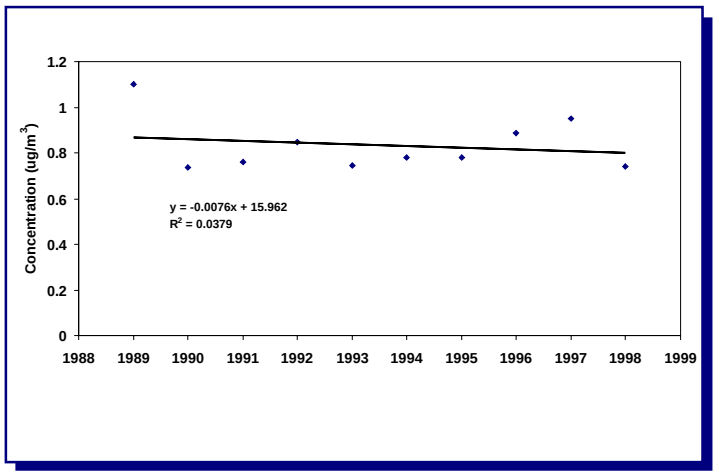


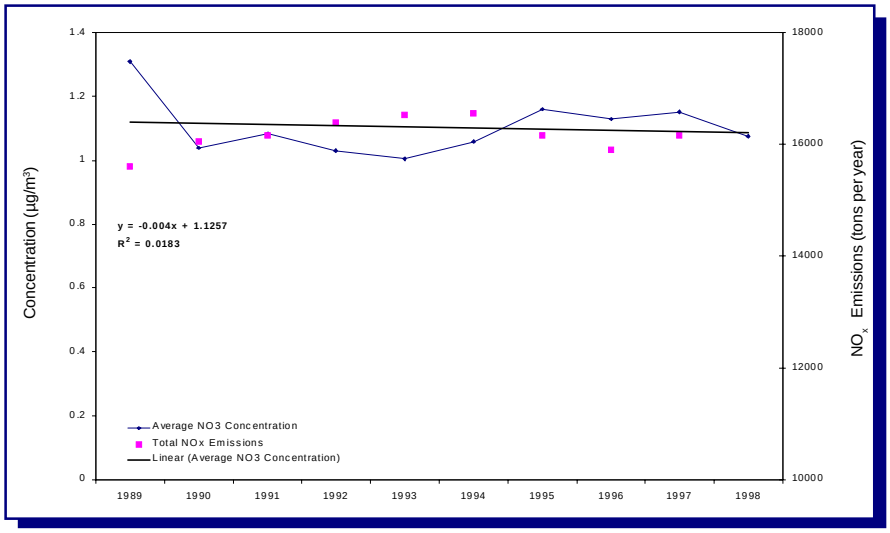
Figure 2-16. Linear Regression Analyses for Annual and Quarterly Average Particulate NO_3^- Concentrations– Eastern United States



First Quarter

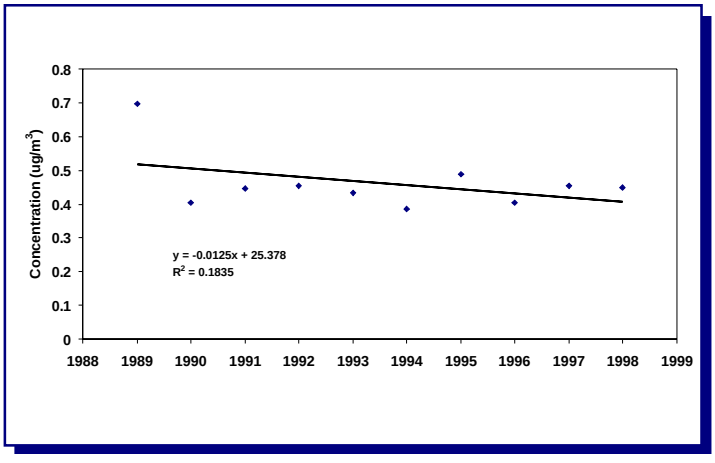


Second Quarter



Annual Average

Third Quarter



Fourth Quarter

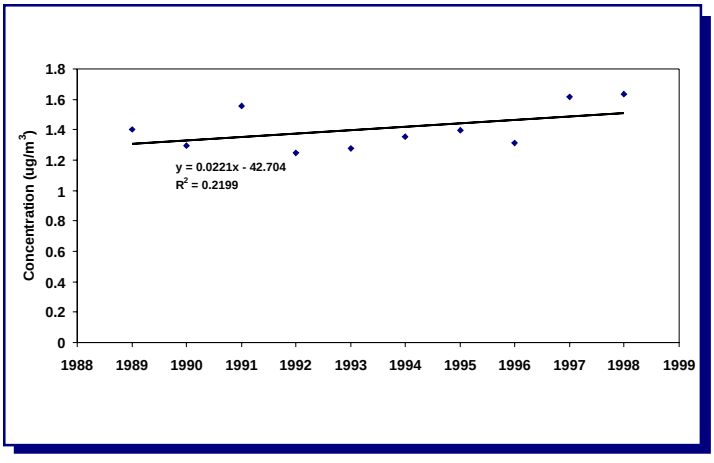


Figure 2-17. Trends in Composite Annual Particulate NO_3^- Concentrations

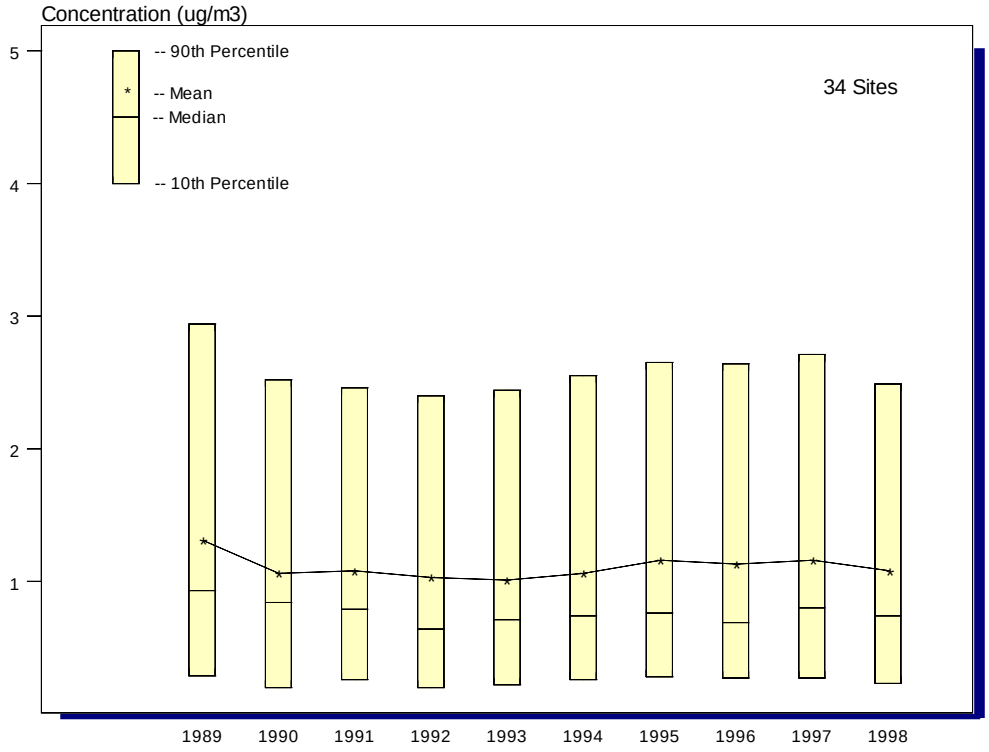
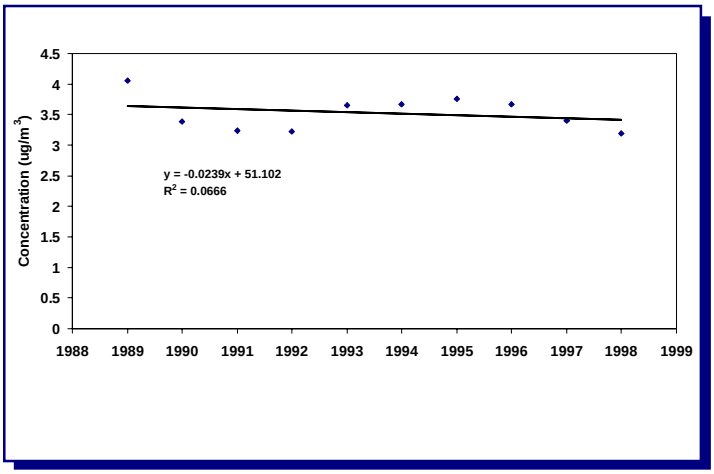
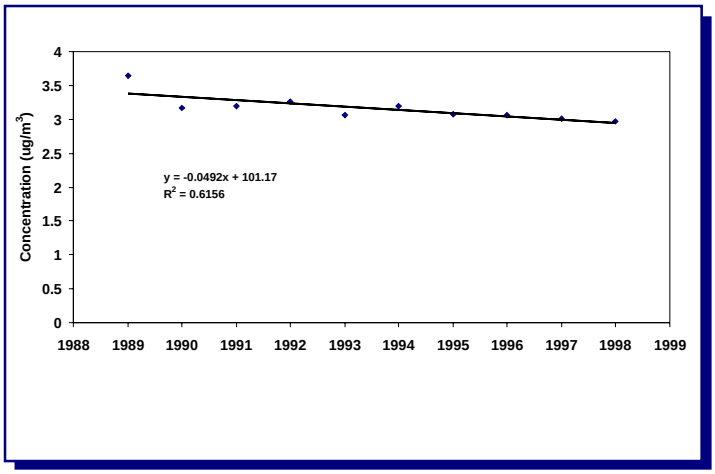


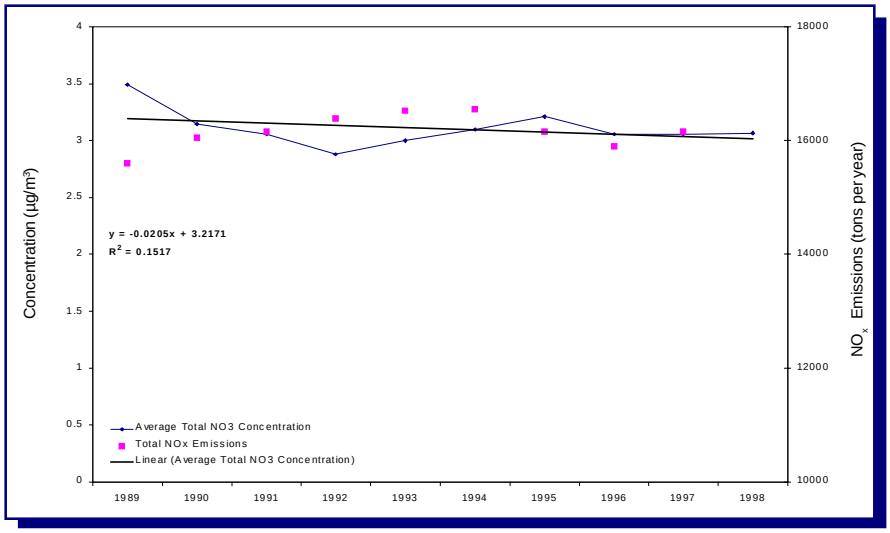
Figure 2-18. Linear Regression Analyses for Annual and Quarterly Average Total NO_3^- Concentrations– Eastern United States



First Quarter

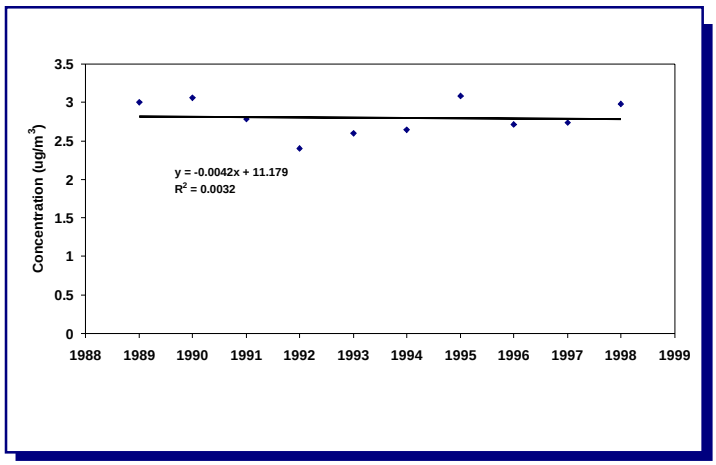


Second Quarter



Annual Average

Third Quarter



Fourth Quarter

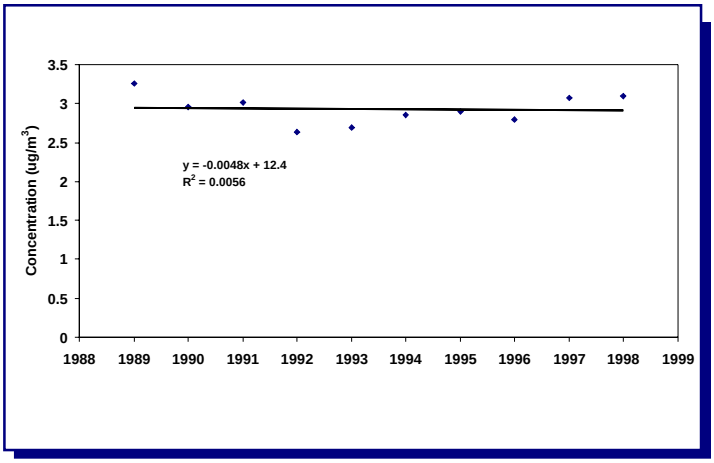
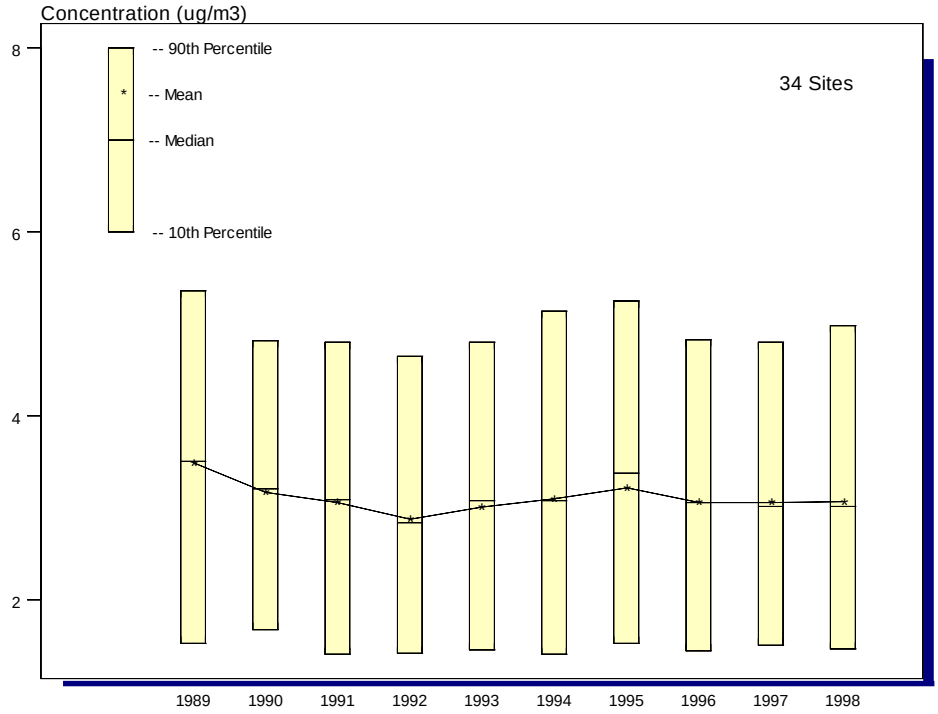



Figure 2-19. Trends in Composite Annual Total NO_3^- Concentrations